Auxiliary Materials for Anisotropic superconductivity in layered $LaSb_2$: the role of phase fluctuations

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We present here plots of the imaginary part of the ac magnetic susceptibility of $LaSb_2$ as function of temperature, and magnetic field for two representative crystals in Fig. 1. Data are shown for two different magnetic field orientations to display the anisotropic behavior.

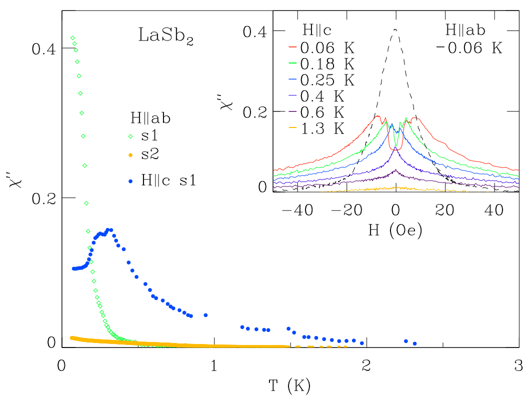

FIG. 1: Ambient pressure superconductivity. Imaginary, part of the ac magnetic susceptibility, χ'', for excitation fields along the c-axis and in the ab plane vs temperature, T, for two representative crystals, s1 and s2. Inset: χ'' for s1 vs magnetic field, H, at T's identified in the figure.

In Fig. 2 we present the anisotrpy in the critical fields at 1.78 K as a function of applied hydrostatic pressure, P as determined by the real part of the ac magnetic susceptibility for a crystal that was nominally aligned ($\pm 10^o$) to the applied magnetic field. We observe a reduction of the anisotrpy with P including isotropic behavior near 6 kbar.

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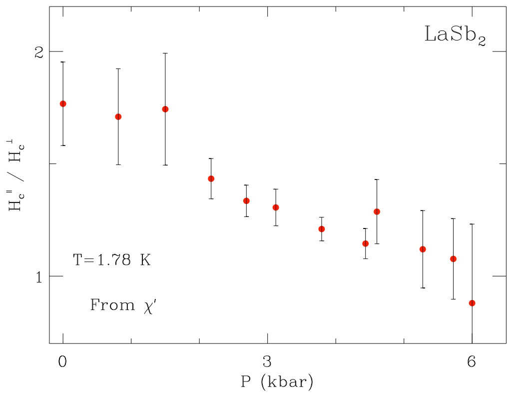

FIG. 2: Critical field anisotropy. Anisotropy of the critical field, H_c, for $H \parallel$ ab planes, $H_c^{\parallel}$, divided by that for $H \parallel$ c-axis, $H_c^{\perp}$, vs pressure, P, as determined by the real part of the ac susceptibility at 1.78 K.

Anisotropic superconductivity in layered $LaSb_2$: the role of phase fluctuations

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We present electrical transport and magnetic susceptibility measurements of the highly anisotropic compound $LaSb_2$ observing a very broad transition into a clean, consistent with type-I, superconducting state with distinct features of 2 dimensionality. Application of hydrostatic pressure induces a 2- to 3-dimensional crossover evidenced by a reduced anisotropy and transition width. The superconducting transition appears phase fluctuation limited at ambient pressure with fluctuations observed for temperatures greater than 8 times the superconducting critical temperature.

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Superconductivity in reduced dimensions has intrigued condensed matter physicists for over 40 years. Highly anisotropic materials with superconducting (SC) phases, such as TaS_2 and $NbSe_2$[1–4], as well as thin SC metallic films[5–7] and organic compounds[8] were investigated to search for novel properties stemming from dimensionality effects. More recent discoveries of unconventional superconductivity in layered cuprates[9], MgB_2[10], and iron pnictides[11], all possessing anisotropic crystal structures, has highlighted the importance of the layered structure and phase fluctuations[12, 13] in determining the SC and normal properties of these compounds. Here we present resistivity, magnetization, and AC susceptibility measurements on the highly layered SC compound $LaSb_2$[14–16] which has been of interest because of its magnetoresistive properties[17]. Previous investigations of $LaSb_2$ show no indication of competing order such as a charge density wave transition[16]. We present evidence that the ambient pressure SC phase, in which only a minority of crystals display a complete Meissner effect at low temperature, T, is characteristic of poorly coupled two dimensional (2D) SC planes. The anisotropy is reduced and the transition is dramatically sharpened as pressure is applied indicating a crossover from a 2D to a more traditional 3D SC phase. Our data demonstrate that the extraordinarily wide, and many times incomplete, SC transition at ambient pressure likely results from 2D phase fluctuations which persist for temperatures much lower than the onset temperature for superconductivity, T_{onset}, that is at T's an order of magnitude larger than the global SC critical temperature, T_c. This places $LaSb_2$ among a handful of systems[7, 13] exhibiting phase fluctuation limited superconductivity and is unusual in that it displays behavior consistent with clean, type I, superconductivity[18].

$LaSb_2$ is a member of the RSb_2 (R=La-Nd, Sm) family of compounds that all form in the orthorhombic, highly layered, $SmSb_2$ structure[14, 17, 19] in which alternating La/Sb layers and 2D rectangular sheets of Sb atoms are stacked along the c-axis. These structural characteristics give rise to the anisotropic physical properties observed in all the compounds in the RSb_2 series[14, 15, 17]. A large number of single crystals of $LaSb_2$ grown from high purity La and Sb by the metallic flux method were large flat, micaceous, plates, which are malleable and easily cleaved. Polycrystalline samples grown in crucibles using a stoichiometric mixture of the constituents had T_{onset} essentially identical to the crystals. The $SmSb_2$ structure-type with lattice constants of $a = 0.6219$, $b = 0.6278$, and $c = 1.846$ nm, was confirmed by single crystal X-ray diffraction. Resistivity, ρ, measurements were performed with currents either in the ab-plane or along the c-axis using standard 4-probe ac techniques at 17 or 27 Hz from $0.05 \leq T \leq 300$ K. Data presented here are from single crystal samples with residual resistance ratios of 70-90 between 300 and 4 K. Magnetization, M, and susceptibility, χ, were measured with a commercial SQUID magnetometer for $T > 1.75$ K and a dilution refrigerator ac χ probe for $T \geq 50$ mK and were corrected for demagnetization effects based upon crystal dimensions. Our ac susceptibility measurements were found to be free of Eddy currents effects as our measurements were independent of excitation frequency and amplitude in the range of parameters employed. The susceptibility of several crystals was measured in the SQUID magnetometer with applied hydrostatic pressure, P, of up to 6.5 kbar in a beryllium-copper cell previously described[20].

Shown in Fig. 1a is ρ measured with the current in the ab plane, ρ_{ab}, and along the the c-axis, ρ_c, of $LaSb_2$ as a function of T in zero magnetic field, H. Note that the normal state ρ is highly anisotropic with $\rho_{ab} = 1.2$ $\mu\Omega$ cm at 4 K and $\rho_c/\rho_{ab} \sim 200$. The ρ_{ab} data suggest a broad SC transition with an onset apparent near $T_{onset} \sim 1.7$ K. However, a true $\rho = 0$ state is not reached until 0.7 K. In contrast, the T dependence of ρ_c indicates an onset near 1.0 K followed by a $\rho = 0$ state below 0.5 K. Interestingly the ρ_c curve also shows a small peak for $T < T_{onset}$ similar to was has been reported in (LaSe)1.14(NbSe2)[21] and attributed to a quasiparticle tunneling channel in the interlayer transport. All of these features can be suppressed with the application of magnetic fields as demonstrated in Fig. 1b where a com-

pelling difference in ρ_{ab} and ρ_c with H oriented along the ab planes is displayed. We observe that a field of ~ 500 Oe completely destroys the SC currents along c-axis while their counterparts in the ab planes remain intact. This demonstrates a relatively poor coupling between the SC condensate residing on neighboring Sb planes. We believe, in fact, that these data represent two transitions: a planar superconducting transition initiating at T_{onset} and a secondary bulk transition below $T_c = 0.5$ K associated with the emergence of coherent interlayer coupling.

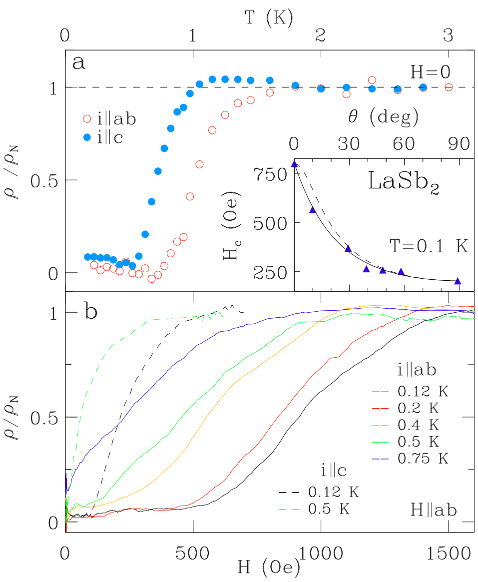

FIG. 1: Resistivity. (a) Resistivity, ρ, divided by the normal state resistivity, ρ_N, vs temperature T for currents along the ab-plane and the c-axis. Inset: Critical field, H_c vs angle, θ, from H parallel to the ab planes at 0.1 K. Solid (dashed) line is a plot of the 2D (anisotropic 3D) Tinkham formula[22, 23]. (b) ρ/ρ_N vs magnetic field, H, in the ab plane for currents perpendicular to H in plane and along the c-axis.

Similar features are observed in the magnetic response of the SC phase in $LaSb_2$, Fig. 2. However because χ and M are more representative of the true thermodynamic state of the system the fragility of this phase results in a high sensitivity to growth conditions, magnetic fields, and, as we show later, P. Although all crystals measured, more than 20, displayed $2.25 \le T_{onset} \le 2.5$ K in χ (Fig. 2a upper inset), a broad range of behavior was found in $\chi(T)$ with an incomplete Meissner effect observed in most crystals. We believe that this sensitivity is due to the proximity of $LaSb_2$ to a fully 3D SC phase. This disparate behavior is demonstrated in Fig. 2a where the real part of the ac χ, χ', is plotted for two of the 3 crystals whose χ was explored at dilution refrigerator temperatures. One crystal, sample s1, displays a very broad transition to a $\chi' = -1$ state at $T < 0.2$ K for ac excitation fields, H_{ac}, oriented along the c-axis. For H_{ac} oriented along the ab planes the diamagnetic signal remains incomplete for s1, approaching -0.75 at our lowest T, while the second sample, s2, displays only a small diamagnetic signal. The full Meissner state in s1 for $H_{ac} \parallel c$ is only apparent below 0.2 K despite a diamagnetism consistent with type I superconductivity at $T < 2.5$ K as demonstrated in Fig. 2b. Here, similarly large anisotropies are apparent in the magnetic field, H, dependence of M, that faithfully reflect the crystalline structure. The dc H dependence of χ' for s1 in the two field orientations are shown in the lower inset to Fig. 2a at a few Ts. In Figs. 2a and b the small characteristic fields for the destruction of the Meissner state are apparent.

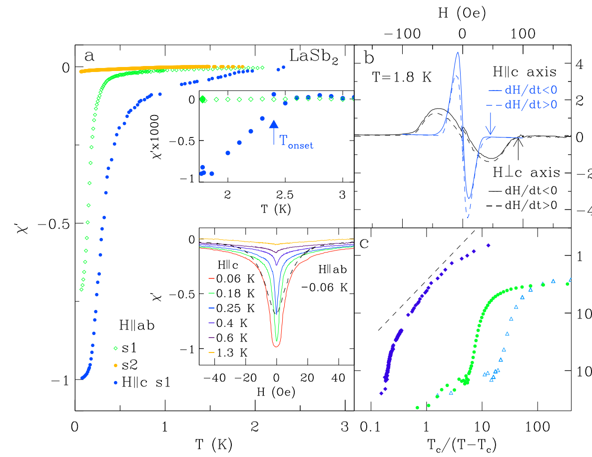

FIG. 2: Ambient pressure susceptibility and magnetization. (a) Real part of the ac susceptibility, χ', for excitation fields along the c-axis and in the ab plane vs temperature, T, for two representative crystals, s1 and s2[23]. Upper inset: detail near the onset of superconductivity, T_{onset} as indicated by the arrow. Lower inset: χ' for s1 vs magnetic field, H, at T's identified in the figure. (b) Magnetization, M, at $T = 1.8$ K vs H along the c-axis and ab planes. (c) $-\chi'/T$ for $H \parallel$ c-axis vs reduced temperature, $T_c/(T-T_c)$ for sample s1 with logarithmic axes at $P = 0$ (blue diamonds) and for a second sample with $P = 2.7$ kbar (green bullets), and $P = 4.4$ kbar (blue triangles). Line is a linear dependence.

Estimates based upon our previous $\rho(T,H)$, Hall effect[17], and de Haas-van Alphen (dHvA)[15] measurements confirm our crystals have small carrier density, n, small carrier mass, m^*, and highly metallic in-plane transport that make anisotropic, type I, superconductivity sensible in $LaSb_2$. The Hall coefficient with $H \parallel c$ is indicative of $n = 2 \times 10^{20}$ cm^{-3}. The small n and low ρ_{ab} indicate highly conductive transport along the ab plane with an estimated Hall mobility of 2.7 m^2/Vs and mean free path, ℓ, of ~ 3.5 μm[17]. The reduction of the dHvA amplitudes with T is small so that m^* is only 0.2 times the bare electron mass[15]. With these parameters, simple estimates[22] of the London penetration depth, λ, and Pippard coherence length, ξ_0, for currents in the ab plane give $\lambda \ge 0.15\mu$m, dependent on the SC condensate fraction, and $\xi_0 = 1.6\mu$m, much larger than

in typical intermetallic compounds. The large ℓ puts our crystals in the clean limit with $\kappa = \lambda/\xi_0 < 1$ consistent with type I superconductivity and a small critical field, H_c. Type I superconductivity is rare in intermetallic compounds and its discovery here is a reflection of the extraordinarily long scattering times for currents in the ab planes[17, 18].

The application of P dramatically reduces the anisotropy and significantly sharpens the transition as we demonstrate in Fig. 3. Here we present the P and T dependence of χ' for Ts near the onset of superconductivity with the same field orientations as in Fig. 2. Although we have only followed χ' down to 1.78 K it is apparent that by 4.4 kbar the transition width has been reduced to ~ 0.1 K with $\chi' = -1$ at 1.8 K for $H_{ac} \parallel c$, while for $H_{ac} \parallel ab$, $\chi' < -0.75$. In addition, we do not observe the sample-to-sample variability that was so apparent in the ambient pressure $\chi'(T)$.

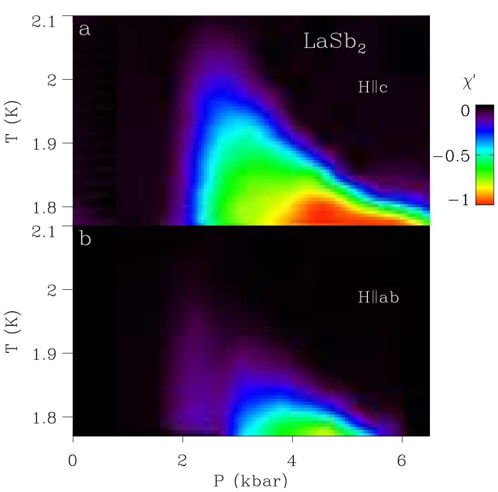

FIG. 3: Pressure and temperature dependence of the superconducting transition. Real part of the ac susceptibility, χ', for magnetic fields H, along the c-axis (a) and along the ab planes (b) vs pressure, P, and temperature, T. These contour plots are produced by simple interpolation of measurements performed at 12 (11) different pressures in frame a (b).

The anisotropies that we measure in ρ and χ' lead naturally to the supposition that the SC in LaSb$_2$ is 2D. To address this possibility we have determined H_c by measuring $\rho(H)$ as a function of field orientation at 0.1 K in the inset to Fig. 1a. We observe a factor of 4 difference in H_c as the crystal is rotated from an orientation where the ab planes are nearly parallel to H ($\theta = 0$), $H_c^{\parallel}$, until they are perpendicular to H ($\theta = 90^o$), $H_c^{\perp}$. For comparison we plot the 2D Tinkham formula[22] prediction, solid line, having no adjustable parameters beyond fixing $H_c^{\parallel}$ and $H_c^{\perp}$ to match our data. The sharp cusp in the data as $\theta \to 0$ is a clear signature of 2D superconductivity. We note that $H_c^{\parallel}$ is much smaller than the paramagnetic limit which has been exceeded in some layered materials[2]. Our measured $H_c^{\parallel}$ is likely intrinsically limited by the long ℓ and large diffusion constant[22], and experimentally limited by the flatness of our crystals.

Our data, thus far, reveal LaSb$_2$ to possess an exceedingly unusual SC phase characterized by large anisotropies for fields and currents parallel and perpendicular to the Sb planes. The SC transition is extraordinarily broad and, in the majority of samples, incomplete at $P = 0$ but is sharpened, and the anisotropy reduced, with application of moderate P. In addition, the SC state at $P = 0$ has an angular dependent H_c characteristic of a 2D superconductor along with features in ρ_c characteristic of quasiparticle tunneling between Sb planes. We believe that the interplane Josephson coupling of essentially 2D SC planes mediates the high pressure 3D phase. There are other mechanisms for these observations that we have considered. The first is the possibility that the SC state at $P = 0$ is restricted to the surfaces of the crystals and that a seemingly unrelated 3D SC state is induced by the application of P. The large Meissner fractions we observe in some of the samples and the continuous evolution of the SC state with P make this very unlikely. Second, we have considered the possibility that we are observing an anisotropic 3D SC state[24] emanating from the 2D-like bands of LaSb$_2$[15]. Anisotropic 3D superconductivity is consistent with the ratio of $H_c^{\parallel}/H_c^{c}$, but not the angular dependence in Fig. 4a. In addition, it is difficult to explain the large anisotropy in ρ and $\chi'(T)$ in Figs. 1 and Fig. 2a in in such a scenario. Finally, we point out that the wide superconducting transition at ambient pressure is not likely caused by impurities or second phases in our crystals since our X-ray diffraction data are free from extraneous peaks, we deduce very long mean free paths for carrier transport along the ab planes, and because the application of moderate pressure is unlikely to suppress the effects of impurities or defects.

Thus, our data suggest that at low T LaSb$_2$ is best described as a set of Josephson coupled 2D planar superconductors. Interestingly, our observation of an extraordinarily wide, and often times incomplete SC transition at $P = 0$, along with the dramatic changes apparent with moderate P, indicate that the SC transition may be limited by phase and amplitude fluctuations of the SC order parameter. Emery and Kivelson have demonstrated that phase fluctuations are dominant when the superfluid density is small so that there is small phase stiffness[12]. Experiments have revealed that the underdoped high T_c SC cuprates are indeed phase fluctuation limited[13]. The importance of phase fluctuations can be determined by a comparison of T_c with the $T = 0$ phase stiffness, $V_0 \propto L/\lambda^2$, which gives the T at which phase order would disappear, T_θ^{max}. Here, L is the characteristic length scale which in quasi-2D superconductors is the larger of the spacing between SC layers or $\sqrt{\pi}\xi_\perp$. With our estimated λ, and with the assumption that

$\xi_\perp < c/2 = 0.93$ nm, we find $T_\theta^{max} \leq 6.1$ times the onset T for superconductivity at ambient pressure (2.5 K), comparable to that tabulated for the cuprates where T_θ^{max}/T_c ranges from 0.7 to 16[12].

One of the consequences of a phase limited transition is an extended T-range where χ' is dominated by fluctuations at $T > T_c$. Ginzburg-Landau (GL) theory, applicable in proximity to T_c, predicts power-law dependencies for χ'/T in the reduced temperature, $t = T_c/(T - T_c)$[22]. To check for such power-laws in the T range over which the SC phase develops we have plotted $-\chi'/T$ as a function of t for s1, where we have used the maximum $\chi''(T)$ to define T_c, in Fig. 2c. The line in this figure represents the form expected in 2D, $\chi'/T \propto t$. The data at ambient P are well described by a power-law form over a decade in t with an exponent that approaches that of the GL 2D prediction. However, the large values of $-\chi'$ that we measure, for example at $t \sim 1$ $-\chi'/T \sim 0.1$, require $\xi_0 \sim 11$ μm, about 7 times the estimate based upon transport data. In contrast, the transitions at $P > 2$ kbar are not well described by a power-law in our range of t as is commonly the case when the SC state has a 3D character and the fluctuation dominated regime is restricted to much larger t. The phase diagram of Fig. 4b demonstrates how this picture evolves with T and P.

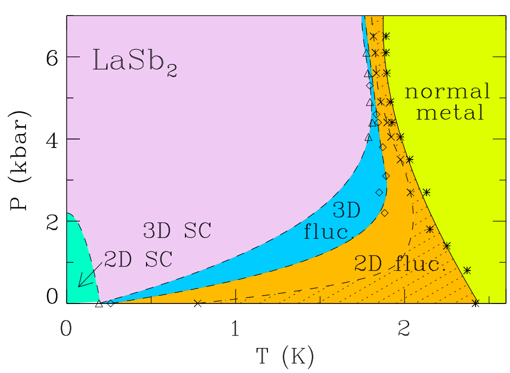

FIG. 4: Phase diagram. Proposed temperature, T, and P, phase diagram. Symbols are onset of diamagnetism (*), 10% (x's) and 90% (triangles) of full Meissner for $H \parallel$ c-axis and 10% of full Meissner $H \parallel$ ab planes (diamonds). Lines are simple interpolations between the data points.

We conclude that at ambient pressure the very anisotropic SC phase of $LaSb_2$ is fluctuation limited with fluctuations extending to Ts an order of magnitude greater than T_c. The small m^*, long ℓ, and small n lead to large in-plane ξ_0 reducing the phase stiffness of the SC state. The application of P increases the Josephson coupling between the SC planes leading to a more traditional isotropic SC transition at the BCS T_c. Thus, our data suggest the existence of a quantum, $T = 0$, phase transition between 2D and 3D superconducting phases with P. In addition $LaSb_2$ is a compelling candidate for investigating the pseudogap region where SC pairs are thought to form at Ts above the phase ordering T, as in the underdoped cuprates, in a BCS superconductor without the complication of a competing ground state.

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